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PHOTOGRAPHIC SPECTROSCOPIC MEASUREMENT OF ULTRAVIOLET SOLID ROC--ETC(U)
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Photographic spectra having a nominal bandpass of 0.4nm were obtained from the radiation from several solid rocket motor exhausts in the spectral region 250nm to 430 nm. The spectra consisted primarily of a continuum with some molecular band structure which was found to originate from the molecules OH, PbO, NO & CHO. These molecular bands were more pronounced in the region 290 nm to 320 nm, but the radiating systems involved extend to shorter wavelengths. The source of the continuum radiation was not determined explicitly, but is not inconsistent witha CO + O recombination mechanism.

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PHOTOGRAPHIC SPECTROSCOPIC MEASUREMENT OF ULTRAVIOLET SOLID ROCKET MOTOR PLUMES

Final Report
By

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February 7, 1980

US Naval Weapons Center
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I. Introduction

The optical signature of a solid propellant rocket motor contains a significant component in the solar-blind ultraviolet region of the spectrum (250 - 300 nm). This radiation is an attractive source for missile detection, since the virtual absence of solar radiation in this region results in very low background noise. The development of solar-blind photomultiplier tubes and optical filters with a sharp low frequency cut-off makes it possible to develop sensitive warning detectors which are not readily susceptible to false alarms.

The mechanisms responsible for the production of solar-blind radiation in a solid propellent rocket plume are not well understood. Low resolution spectra indicate that the radiation is essentially a continuum, rising in intensity as the wavelength increases from 250 nm to 300 nm in a manner similar to a blackbody with a temperature near 3000°K.

Several mechanisms have been proposed which could contribute to this radiation. One source is radiation from particles in the plume. Since the plume temperature is typically considerably less than 3000°K, the particle radiation would not exhibit the observed spectral distribution unless the particles were at a higher temperature than the plume, or the emissivity decreased with increasing wavelength. The first possibility was discussed by Mann et al [1] who suggested that aluminum oxide particles may be found by sublimation at a temperature higher than that of the plume. The second possibility is suggested by a discussion of the radiation from small particles given by McGregor [2]. Regardless of the details of the mechanism, particle radiation appears to contribute to the

total solar-blind radiation, and it is observed that the intensity increases considerably in those plumes in which the particle density is high.

Another possible source of radiation in the solar-blind ultraviolet region is chemi-luminescence. Molecules formed in the plume in excited electronic states decay to a lower energy state producing radiation characteristic of the molecule.

Several molecules which could be present in rocket plumes have emission bands which lie within the 250 - 300 nm range. The most likely candidates are OH, CO_2 and CHO. Other possible emitters include O_2, NO and CO.

Previous spectral measurements of rocket plumes have employed scanning monochrometers to record the spectrum. Since there are relatively large fluctuations in the overall plume intensity during the duration of a spectral scan, it is difficult to differentiate between intensity fluctuations and spectral features. For this reason, and the necessity to perform a complete spectral scan during the comparatively brief duration of the firing, the effective spectral resolution has been the order of 2 - 5 nm. This is inadequate to resolve the characteristic spectra of the various molecules which are candidate emitters.

Photographic spectra, time integrated over the total firing duration, have been obtained which have an effective bandpass of approximately 0.4 nm. These spectra were obtained during static test firings of several tactical missiles at Allegany Ballistic Laboratory during August, 1979. An earlier attempt to obtain similar spectra during the firing of several model rockets at the Naval Ordinance Station were largely unsuccessful as a result of scattered light from a concurrent ultraviolet scattering experiment and the sun.

The results of these experimental measurements are reported herein. They indicate that in the region 250 - 300 nm the radiation is largely continuous in nature, but there are some contributions from molecular species, particularly at the longer wavelength range. Significant radiation was observed from the previously unreported molecule PbO in several of the missile exhaust plumes. Band radiation from PbO and OH and continuum radiation from CO_2 may contribute to the apparently large radiation temperature in the 250 - 300 nm spectral range.

II. Experimental Procedure

The objectives for this experiment were to obtain spectra with sufficient resolution to determine whether molecular band emission was present in solid propellent rocket plumes in the 250 - 300 nm region and to identify the molecules responsible for the emission. In order to satisy these objectives the highest possible spectral resolution was required. Photographic recording of the spectra was chosen to satisfy the requirement for high resolution.

The intensity of radiation produced by a rocket plume is not constant, but fluctuates in time. In a scanning spectrometer these fluctuations may be interpreted as spectral features. Normalization of the scanned data with respect to the total radiation from the plume is insufficient to remove this uncertainty, since not all spectral regions will share the same temporal fluctuations. With photographic spectra all spectral regions are recorded simultaneously and integrated over the entire firing duration by the photographic process. This insures that relatively narrow spectral features are not masked by intensity fluctuations. Temporal resolution of the data is lost, but this has little effect on species identification.

One drawback of the photographic method is the lack of linearity and relative insensitivity when compared with photoelectric methods. The sensitivity of the film limits the resolution which can be obtained to considerably less than that which is theoritically possible with moderate size spectrographs. Since the firing duration is typically of the order of one second, the slits must be opened wide enough that sufficient energy is admitted to obtain a proper exposure. Even for the relatively sensitive film used for the ABL test series (Kodak Royal X-Pan) the required slit opening effectively limited the bandpass to approximately 0.4 nm.

The spectrometer was used without external optics, and placed as near as possible to the source, to insure that the source filled as much of the instrument field-of-view as possible. This provides the maximum possible exposure.

The experimental layout used for the test series at ABL is shown in Figure 1. The spectrograph was a 1 meter focal length Czerny-Turner instrument manufactured by Jarrell-Ash. Equipped with a grating having 1180 grooves/mm, the nominal dispersion in the 250 - 300 nm range was 0.8 nm/mm. The maximum slit opening of 0.5 nm was used resulting in a nominal bandpass of 0.4 nm.

The nominal aperture of the instrument is f/8 but the field-of-view is determined by the rectangular grating. For the test series at ABL the angle between the plume axis and the line-of-sight was 63.1°, and the entrance slit was placed 246 cm from the intersection of the plume axis and the line-of-sight. This instrument placement resulted in a field-of-view which was 26.5 cm x 26.5 cm at the plume axis.

Alignment of the spectrometer field-of-view was insured by use of a He-Ne laser. The laser beam was expanded and passed through the exit slit of the spectrometer to overfill the spectrometer optics. The wavelength was set to 632.8 nm and the beam passing out of the entrance slit defined the field-of-view at the plume axis.

The photographic spectra were recorded on Kodak Royal X-Pan, 4" x 5" estar base film sheets. Prior to exposure to the emission from the rocket plume, three calibration exposures were given to each film sheet. The first of these was a strip exposed through a calibrated step density filter with light from a photographic enlarger. This permits determination of the H-D curve for the film for each exposure. The second exposure was a strip exposed to a calibrated deuterium lamp with the line spectrum from a low pressure mercury lamp superimposed. This procedure insures that both wavelength and relative intensity of the rocket plume spectra can be determined from the developed film sheet.

Once exposed, the film sheet was taken to the photographic laboratory at ABL and developed for 8 minutes in Kodak D-11 developer followed by 30 seconds in a stop bath and 8 minutes in Kodak fixer. The developed film sheets were then washed and allowed to air dry.

During the first test series at NOS it was noted that for spectral regions of wavelength greater than the solar blind limit (> 300 nm) the spectra were contaminated by radiation scattered from the rocket plume or the background. This precluded the use of these spectral regions for species identification. One exception was recorded early in the morning on a heavily overcast day (Test 1-C). Further,

within the solar blind region (< 300 nm) the spectra were contaminated by radiation scattered by the plume from a 200 watt mercury-xenon lamp being used as a source, in another experiment, for simultaneous transmission measurements.

In order to avoid these complications the ABL tests were, at our request, performed at night, and all light sources having the capability of emitting in the ultraviolet region of the spectrum were omitted during this test series. Therefore, the results presented in the next section were obtained from spectra which truly represent the emission from the rocket plume, free from any extraneous sources.

III. Results

Each film strip obtained during the test series at ABL and test 1-C at NOL was examined in an attempt to determine whether any molecular species contributed to the radiation. Each spectrum covered approximately 60 nm. For those missiles of which duplicates were fired, the spectrum for the first missile fired covered the range 250 nm to 310 nm, and for the second missile fired the spectral range was 310 nm to 370 nm. In one instance it was possible to obtain a spectrum for the range 370 nm to 430 nm.

A Jarrell-Ash scanning densitometer was used to record the film density onto a paper strip chart for each spectrum obtained. These records were examined to determine the molecular species present. Figure 2 shows a densitometer record for test 1-C, obtained at NOS. Although it is the only usable record from the NOS test series, it is typical of many of the spectra obtained at ABL.

The identification of molecular species from photographic spectra such as these is, necessarily, somewhat subjective. Positive identification with a large degree of confidence can be made only if several different spectral features (vibrational bands) of the molecule can be found. The relative intensity and shading of these bands should correlate reasonably well with other reported spectra of the candidate molecule if a positive identification is to be made.

In the region 250 - 310 nm the radiation consists of a continuum whose intensity increases rapidly with wavelength. Any molecular structure is superimposed on this continuum and relative intensities are difficult to determine. This is particularly true because of the non-linear response of the photographic material. The variation in intensity of the spectrum in this region was usually sufficient to span both the "toe" and "knee" regions of the H-D curve. In the 310 to 370 nm region the continuum radiation is quite strong, but much more uniform in intensity. Therefore, the relative intensities of various molecular features were much easier to determine, and it was often possible to make species identifications with a greater degree of confidence.

Preliminary investigation showed that the following species might be present in some or all of the spectra:

1. OH ($A^2\,\Sigma^+ - X^2\pi$) - This consists of a system of multiple headed bands whose strongest feature is the 0,0 band at 306.4 nm. The system extends from 347 nm down to 244 nm with a relatively strong 1,0 band at 281 nm.

2. PbO - Two systems are found in the region 250 - 370 nm. These are: System D, 359 - 321 nm, whose

strongest features are the 2,3 band at 348.5 nm and the 0,1 band at 340.2 nm, and System E, 306 - 278 nm, whose strongest feature is the 2,1 band at 286.6 nm.

3. NO - Two systems occur in the 250 - 370 nm region. the β system is an extensive system of double headed bands degraded to the red extending from 243 - 527 nm. The δ system is a system of double headed bands degraded to the violet which extends from 196 - 346 nm.

4. CHO- A system of red degraded bands extending from 242 - 409 nm. The most intensive bands occur in the 270 - 350 nm region.

5. CO_2- The spectrum of CO-O_2 flames exhibits a band structure superimposed on a recombination continuum. The bands are relatively indistinct for normal flames at atmospheric pressure. The continuum intensity peaks at approximately 430 nm and extends below 300 nm.

Each of the spectra were examined to determine if the species listed above were present. In light of the subjective nature of this determination the probability that the spectrum contained evidence of a particular species was assigned as follows:

1. Species which are designated 1 have a high probability of being present. This category is applied only if several bands of a system are distinct, have proper shading and have a reasonable relative intensity correlation.

2. Species which are designated 2 are probably present. This category is applied when there are two or more features present at the proper wavelength, but which may not have a strong relative intensity correlation.

3. Species which are designated 3 are considered to be possible. This is an indication that at least one spectral feature appears at the proper wavelength.

Each spectrum was examined and the probability that a species was present was assigned using the criteria described above. The results appear in Table I. From the entries in the table it is evident that OH is present in most of the spectra in the range 250 - 310 nm and many of the spectra in the 310 - 370 nm region. There is strong radiation from PbO in several spectra in the range 310 - 370 nm and it is probably in several of the spectra in the 250 - 310 nm region. None of the other species appear strong enough to make a positive identification. The frequency with which NO was judged probable, suggests that it is present in at least some of the tests. The lack of a positive identification of CO_2 from its band spectrum is not surprising in light of the fact that the radiation is dominated by the continuum at atmospheric pressure.

In the one spectrum taken in the wavelength region 370 - 430 nm the film was, unfortunately, heavily exposed. However, the intensity increased with wavelength up to 430 nm as would be expected for the CO + O recombination continuum. There was no evidence of molecular band radiation in this spectrum.

The dominant characteristic of the radiation in the 250 - 300 nm spectral region is a continuum which increases

rapidly with wavelength. This radiation is probably due to a combination of particle radiation, CO + O recombination and contributions from OH and PbO chemiluminescence. The contributions from the molecular species are strongest at the long wavelength end of this range and would make the increase in total radiation with wavelength even more pronounced. This may explain the fact that graybody radiation temperatures for this spectral region often exceed the plume temperature.

IV. Summary and Conclusions

Photographic spectra having a nominal bandpass of 0.4 nm were obtained from the radiation from several solid rocket motor exhausts in the spectral region 250 nm to 430 nm. The spectra consisted primarily of a continuum with some molecular band structure which was found to originate from the molecules OH, PbO, NO and CHO. These molecular bands were more pronounced in the region 290 nm to 320 nm, but the radiating systems involved extend to shorter wavelengths. The source of the continuum radiation was not determined explicitly, but is not inconsistent with a CO + O recombination mechanism combined with thermal emission from particulates.

If further spectroscopic measurements are made, we recommend that the region 300 - 500 nm be given particular attention to determine whether the continuum radiation exhibits the peak near 430 nm characteristic of the CO + O continuum.

REFERENCES

(1) D. M. Mann, T. D. McCay, E. Steinbrenner and W. A. Danne, "Initial In Situ Plume Property Measurements In a SRM Exhaust", 11th JANNAF Exhaust Plume Technology Meeting.

(2) W. K. McGregor, "On the Radiation From Small Particles", J-Wuant. Spectrosc. Radiat. Transfer, 19, 659, 664 (1978).

(3) R. W. B. Pearse and A. G. Gaydon, The Identification of Molecular Spectra, 3rd Ed., Chapman and Hall Limited, London (1963).

TABLE I

TEST NO.	WAVELENGTH (nm)	OH	PbO	NO	CHO	Pb
27491	250 - 310	1	2	3		
27492	310 - 370		3			
27493	250 - 310	3	3			
27494	310 - 370		2			X*
27495	370 - 430	strong continuum no structure				X*
27496	250 - 310	strong continuum no structure				X*
27497	250 - 310	strong continuum no structure				X*
27498	250 - 310	1	2	3	3	
27499	310 - 370		1		3	
27500	250 - 310	1		3	2	
27501	310 - 370		1	2		X*
27502	250 - 310	1	2		3	
27503	310 - 370		1	2		X*
27504	250 - 310	1	3		3	
27506	310 - 370	3	1	2		X*
27507	250 - 310	1	2	2		
27508	310 - 370	3	1	3		X*
27509	250 - 310	1		2	2	
27510	310 - 370	2	2	3		
1-C	300 - 332	2	1	2	3	

* The sumbol X indicates that emission lines from the atomic spectrum of lead (PbI) were observed. The observed lines were of wavelength 368.3, 364.0 and 282.3 nm.

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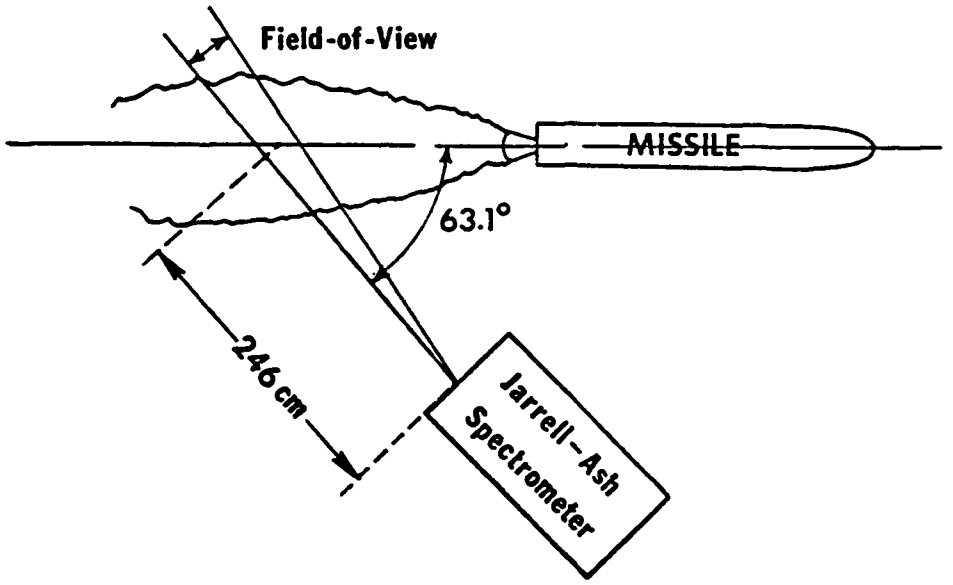

FIGURE 1. Experimental configuration for the tests at ABL.

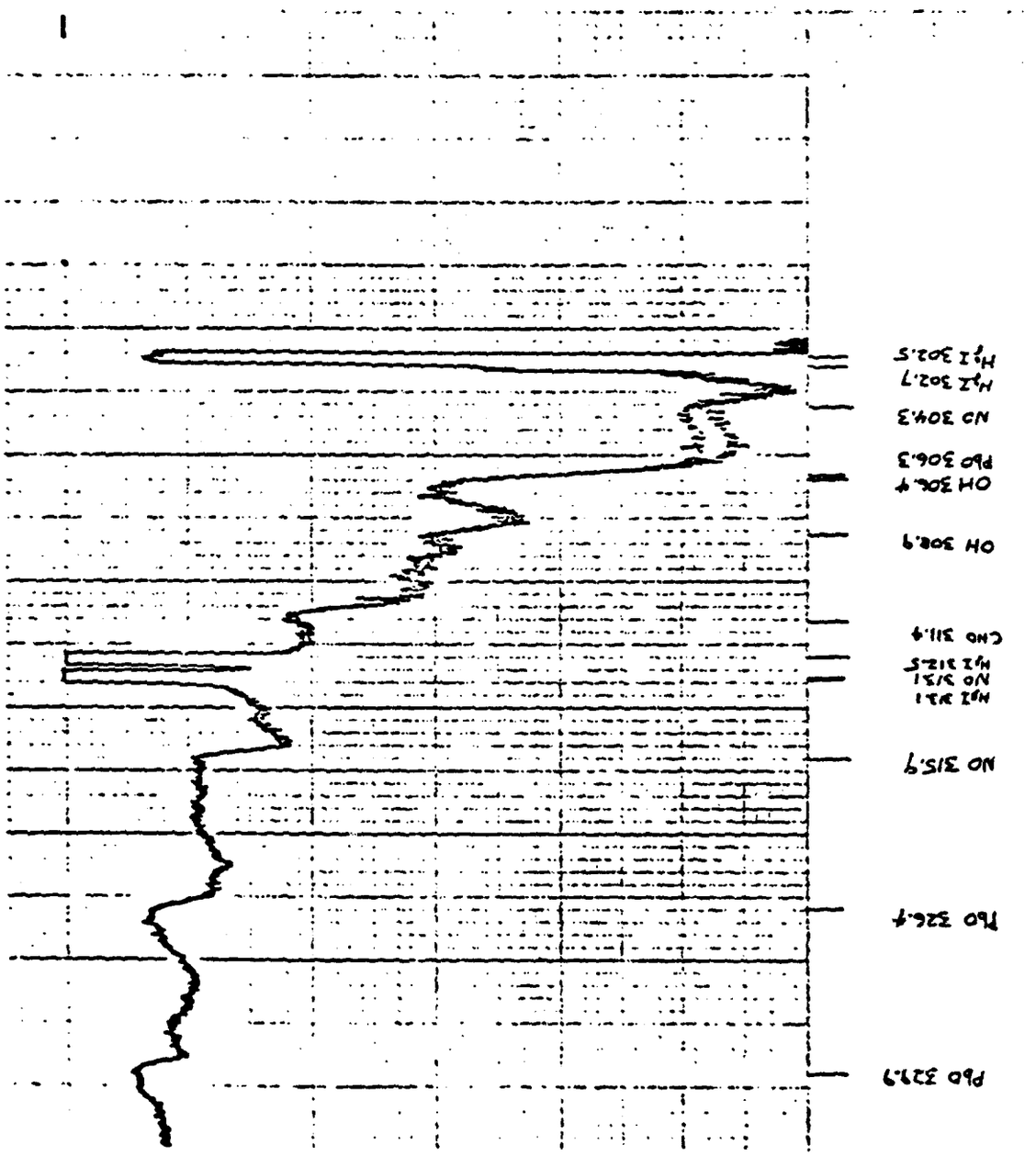

FIGURE 2. Densitometer scan of the photographic spectrum of test 1-C obtained at NOL. The spectral features which were identified are shown below together with their wavelength. The sharp intense lines belong to the superimposed mercury spectrum used for wavelength calibration.